



# A novel bipolar charger for submicron aerosol particles using carbon fiber ionizers

Bangwoo Han<sup>a,b</sup>, Neelakshi Hudda<sup>a</sup>, Zhi Ning<sup>a</sup>, Hak-Joon Kim<sup>b</sup>, Yong-Jin Kim<sup>b,\*</sup>,  
Constantinos Sioutas<sup>a,\*</sup>

<sup>a</sup>Department of Civil and Environmental Engineering, University of Southern California, Los Angeles, CA 90089, USA

<sup>b</sup>Eco-Machinery Research Division, Korea Institute of Machinery and Materials, Daejeon 305-343, South Korea

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## ABSTRACT

A simple and novel bipolar charging device using carbon fiber ionizers was developed to neutralize submicron aerosol particles without the generation of ozone. The ion currents of the positive and negative ions generated by carbon fiber ionizers were so chosen as to optimize particle neutralization. The particle penetration, charging probability and charge distribution resulting from the charger were investigated and compared to those from a Kr-85 radioactive neutralizer for the particles in the size range of 20–120 nm. Size distributions for various laboratory-generated aerosols (sodium chloride, ammonium nitrate, ammonium sulfate and glutaric acid) neutralized by the charger were also investigated and compared to those obtained without neutralization. Particle penetration in the charger was over 90% for particles larger than 20 nm. Charging probability and charge distribution for the charger were in good agreement with those from Kr-85 neutralizer and with theoretical estimations. Size distributions observed for the charger and Kr-85 neutralizer were also in good agreement for particles of different concentrations and various chemical compositions. The newly developed bipolar carbon fiber charger can neutralize submicron particles, at least as effectively as currently available radioactive neutralizers and with negligible ozone generation which is its major advantage.

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## 1. Introduction

Charging of aerosol particles has become one of the most important tools to control the behavior of particles in aerosol generation, transport and measurement processes. Bipolar charging (i.e. charge neutralization) has been particularly important in aerosol sizing and measurement systems, such as a differential mobility analyzer (DMA; [Knutson & Whitby, 1975](#); [Kousaka, Okuyama, & Adachi, 1985](#)) and particle beam mass spectrometers ([Ziemann, 1998](#)). Charge neutralization is essential in decreasing the initial electrical charges on the particles produced by aerosol generators and thus reducing the particle deposition in tubes or valves during transport.

Radioactive materials have been commercially used for particle neutralization. Radioactive sources such as Am-241, Kr-85 and Po-210 have been used for the production of stable and equal number of positive and negative ions by means of radioactive decay. However, legal restrictions limit their broader use because of the possibility of radioactive leaks.

\* Corresponding authors. Tel.: +82 42 868 7475, fax: +82 42 868 7284 (Yong-Jin Kim); tel.: +1 213 740 6134, fax: +1 213 744 1426 (Constantinos Sioutas).  
E-mail addresses: [yjkim@kimm.re.kr](mailto:yjkim@kimm.re.kr) (Y.-J. Kim), [sioutas@usc.edu](mailto:sioutas@usc.edu) (C. Sioutas).

Corona discharge has also been explored as a means to neutralize particles, since this technique can easily generate a high number concentration of positive and negative ions and thus neutralize aerosols at higher flow rates and concentrations (Adachi, Pui, & Liu, 1993; Romay, Liu, & Pui, 1994). Nonetheless, these chargers showed some problems such as imbalance of positive and negative ion concentrations, particle generation by sputtering of the electrodes or by chemical reaction as well as ozone emission during corona discharging (Murray & Gross, 1989; Romay et al., 1994; Sakata, Inaba, Yoshida, & Okada, 1991). Ozone generated in the corona discharger is of particular concern, as it may alter the chemical and microphysical properties of the charged particles by means of chemical reactions involving particle-bound organic components, such as polycyclic aromatic hydrocarbons and alkanes (Eliaison, Aloisio, Donaldson, Cziczo, & Vaida, 2003; Katrib et al., 2004).

As an alternative to radioactive sources and corona discharge, soft X-ray has been attempted as a particle neutralization device (Shimada, Han, Okuyama, & Otani, 2002). The soft X-ray charger can generate a high number of stable ions and thus bring particles into the stationary charge states in shorter charging time than the radioactive chargers. However, they utilize an expensive and complicated soft X-ray generator and controller. A novel corona discharger for charging submicron particles using high frequency AC voltage with a capacitor was recently developed (Stommel & Riebel, 2004). This discharger proved to be an attractive alternative to radioactive source with low particles losses. However, it resulted in generating 100–1200 ppb ozone concentration under typical operation conditions, a range that exceeds the National Ambient Air Quality Standards (NAAQS) of US for ozone (80 ppb). More recently, a surface-discharge microplasma aerosol charger (SMAC) was developed for neutralizing aerosol particles (Kwon, Sakurai, Seto, & Kim, 2006). The SMAC had minimum particle losses and showed good potential for replacing traditional radioactive sources. However, the surface-discharge plasma neutralizer still generated ozone in the range of about 100 ppb.

Carbon fiber ionizers can be simply and inexpensively developed by applying a few kV to a bundle of carbon fibers to generate positive or negative ions. They can produce stable ions with sufficiently high concentrations, without generating ozone and thus, they have been used in indoor air purifiers to generate negative ions as an alternative to corona discharge (Chen, Huang, Lin, Chen, & Hsu, 2006). We have found that carbon fiber ionizers can charge fine and ultra-fine particles with low particle losses and as effectively as existing corona chargers, while generating negligible ozone and other particles (Han, Kim, Kim, & Sioutas, 2008). This technique can be also easily applied to generate high number concentrations of stable bipolar ions for neutralizing aerosols. To the best of our knowledge, no previous attempts were made to study bipolar charging of submicron aerosol particles using these carbon fiber ionizers.

In the present work, we describe the development of a novel bipolar charging device using carbon fiber ionizers and discuss its performance characteristics, including particle penetration, charging probability, charge distribution and size distribution. Charging probability and charge distribution resulting from the bipolar charger were compared to those obtained from a radioactive neutralizer as well as theoretical predictions based on diffusion charging theory.

## 2. Experimental setup

Fig. 1 shows the carbon fiber bipolar charger developed in this study which consists of two carbon fiber ionizers and a charging chamber. Each carbon fiber ionizer consists of a carbon fiber electrode placed inside a grounded stainless steel (SS) cylinder. The carbon fiber electrode is a bundle of approximately 300 carbon fibers, each about 5–10  $\mu\text{m}$  in diameter and 5 mm in length; this is the same type of carbon fiber electrodes used in indoor air purifiers. The bundle of carbon fibers is connected to a 1/8" SS rod with a crimp socket connector and then covered with a heat shrinkable tube. The SS rod is covered with a silicon tube inside a SS cylinder for electrical insulation. Compressed filtered air is introduced at 4–6 l/min into the SS cylinder via a 1/4" SS tube to drive the ions generated in the ionizer into the charging chamber. The end of the SS tube is placed near the tip of carbon fiber to create a jet that flushes out the generated ions to the charging chamber, thus minimizing the loss of ions in the SS cylinder where a high electric field is formed. Two ion streams of positive and negative ions created by the carbon fiber ionizers operating at positive and negative DC voltage of 2.0–4.0 kV, respectively, are introduced into the charging chamber. The aerosol stream is introduced into the charging chamber at 1.5 l/min and mixed with the incoming ion streams. The residence time of the particles is about 1.0 s in the chamber, which has a volume of 188  $\text{cm}^3$ .

Fig. 2 shows the experimental setup for measuring the charge probability, charge distribution and size distribution of particles in the bipolar charger. Sodium chloride (NaCl) aerosol particles within the size range of 10–500 nm were generated by nebulizing NaCl solutions with a nebulizer (VORTAN Medical Technology, Inc., Sacramento, CA) and mixed with particle-free filtered air and then passed through a dilution chamber to reduce the particle concentration to within the operating range of measurement devices, such as DMA and condensation particle counter (CPC). Total number concentration of the particles was monitored by a CPC (Model 3022A, TSI Inc., St Paul, MN) at the exit from the dilution chamber. Particles were then passed through a Kr-85 neutralizer (Model 3012, TSI Inc. St. Paul, MN) at a rate of 1.5 l/min before being introduced into a DMA (1st DMA; Model 3071, TSI Inc., St. Paul, MN) with a sheath flow of 15 l/min. The particles exiting the 1st DMA, at a given voltage, were introduced into another Kr-85 neutralizer, followed by a parallel plate electrostatic precipitator (1st ESP) to which a DC high voltage of about 10 kV is applied. The particles exiting the 1st ESP can be considered as nearly non-charged monodisperse particles. The fraction of multiply charged larger particles was reduced to about or less than 10% by carefully adjusting the size distribution of the test aerosol, as we have discussed in our paper, Han et al. (2008). The number concentration of the generated monodisperse particles was  $7.1 \times 10^2$ ,  $7.6 \times 10^3$ ,  $1.1 \times 10^4$ ,  $7.5 \times 10^3$ ,  $6.1 \times 10^3$  and  $4.1 \times 10^3/\text{cm}^3$  for 20, 40, 60, 80, 100 and 120 nm, respectively. The

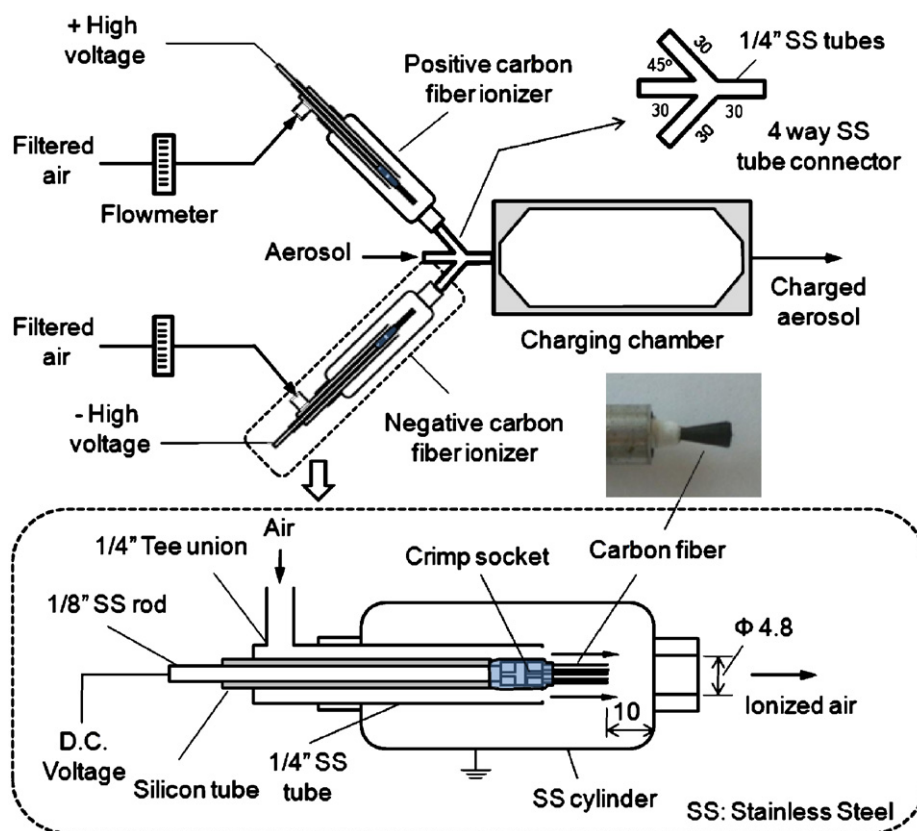


Fig. 1. Schematic of the carbon fiber bipolar charger developed in this study.

monodisperse aerosol was used for measuring the charging probability and charge distribution for particles of different sizes. This test aerosol was introduced into the carbon fiber bipolar charger (or the other Kr-85 neutralizer for comparison) and then drawn through a second ESP (2nd ESP) and a CPC (Model 3022A, TSI Inc., St. Paul, MN) for measurements of charging probability. Particle concentrations were measured without applying a voltage to the 2nd ESP ( $n_1$ ) and with an applied voltage to the 2nd ESP ( $n_2$ ). The charging probability for the particles can be expressed as  $1 - n_2/n_1$ . The test aerosols charged by the bipolar charger or the Kr-85 neutralizer were further drawn into the 2nd DMA and a CPC for measuring charge distribution. External positive and negative voltages were applied to the 2nd DMA to obtain the charge distribution of negatively and positively charged particles, respectively. Ion currents were measured by an aerosol electrometer (Model 3068B, TSI Inc., St. Paul, MN) at the outlet of the carbon fiber charger to monitor the net charge generated by the charger during the experiments. For measuring size distribution, polydisperse NaCl particles generated by the nebulizer were directly introduced into the bipolar charger (or the Kr-neutralizer for comparison tests) and then to the 2nd DMA and CPC. The TSI SMPS software (Aerosol Instrument Manager, Version 8.0) was used to calculate particle size distribution from the particle mobility distributions of the polydisperse aerosols measured with the DMA–CPC system. Various particles of different chemical compositions (ammonium nitrate, ammonium sulfate and glutaric acid) were tested to verify the applicability of the bipolar charger as a neutralizer to atmospheric aerosols. Ammonium nitrate and ammonium sulfate were selected as test aerosols because they represent the two most predominant inorganic salts in ambient PM 2.5 (Malm, Schichtel, Pitchford, Ashbaugh, & Eldred, 2004; Sardar, Fine, Mayo, & Sioutas, 2005). Glutaric acid is a dicarboxylic acid found in ambient aerosols and was selected to represent a typical product of secondary aerosol formation by photo-oxidation of organic gaseous precursors (Cruz & Pandis, 1999; Sempere & Kawamura, 1994). For comparison tests by the Kr-85 neutralizer, a dilution air flow of 10 l/min was introduced and exited in front of the Kr-85 neutralizer to obtain the same particle concentrations as those in the carbon fiber charger. Because one radioactive source sometimes is not sufficiently strong to neutralize charged particles (Covert, Wiedensohler, & Russell, 1997; Stommel & Riebel, 2004), to determine whether our Kr-85 neutralizer could bring the polydisperse particles to the stationary charging state, an additional Kr-85 was connected in series in front of the DMA–CPC and size distributions were measured with one or two Kr-85 neutralizers, respectively. No differences in the aerosol size distributions were found between one and two radioactive sources preceding the DMA, which indicates that one Kr-85 neutralizer is sufficient to bring the polydisperse particles to the stationary charging state.

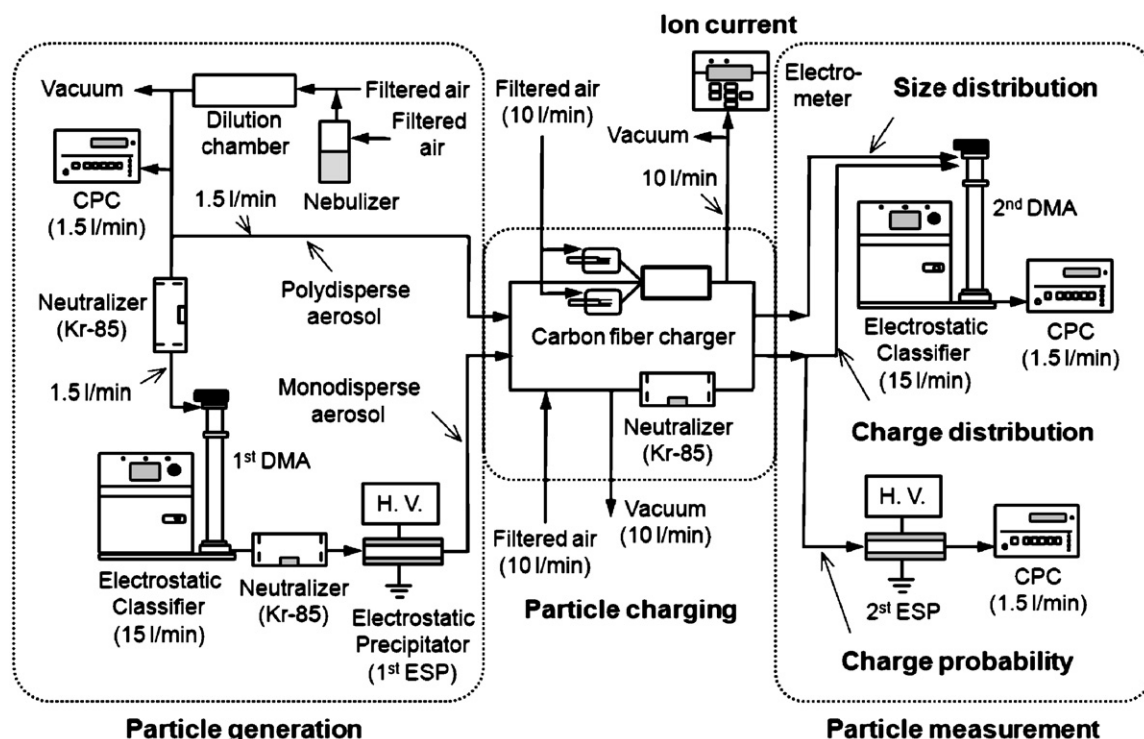


Fig. 2. Schematic of the experimental setup for the measurements of charge probability, charge distribution and particle size distribution.

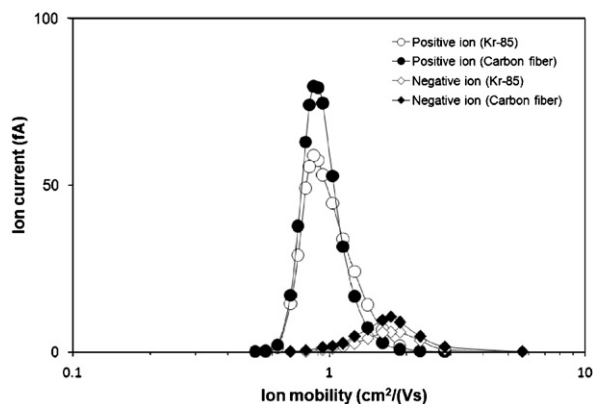
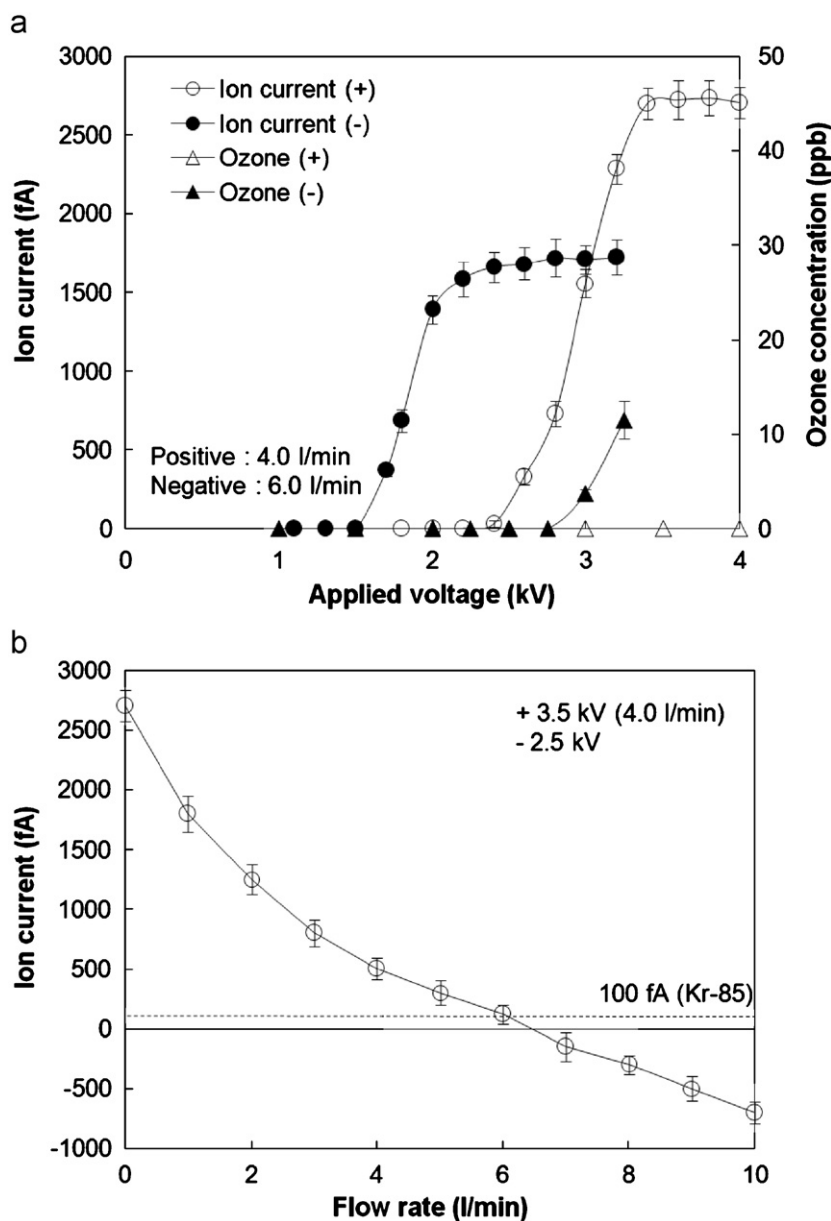


Fig. 3. Mobility distributions of positive and negative ions generated by the carbon fiber ionizer and Kr-85 neutralizer.

### 3. Results and discussion

#### 3.1. Ion mobility distribution

Mobility distributions for the positive and negative ions generated by the carbon fiber charger and the Kr-85 neutralizer are shown in Fig. 3. HEPA-filtered room air (temperature : ~23 °C, humidity : 40–60%) of 4 l/min (or 6 l/min) was introduced into the positive (or negative) carbon fiber ionizer, to which +3.5 kV (or –2.5 kV) was applied and filtered room air of 2 l/min was introduced into the Kr-85 neutralizer. The generated ion stream of 2 l/min was drawn into a nano-DMA (Model 3085, TSI Inc., St. Paul, MN) (with positive and negative applied voltages of 0–10 V) and an aerosol electrometer. The sheath flow rate of the nano-DMA was 20 l/min. Peak values for mobility were observed at about 0.9 and 1.7 cm²/(Vs) for positive and negative ions, respectively, regardless of whether the ions were generated from the bipolar charger or Kr-85 neutralizer. The mobility values for



**Fig. 4.** Ion current of (a) individual positive and negative ions generated by carbon fiber ionizers with different applied voltages and (b) mixed bipolar ions generated by both the positive carbon fiber ionizer (+3.5 kV) with an air flow rate of 4 l/min and the negative carbon fiber ionizer (−2.5 kV) with different air flow rates.

positive and negative ions were similar (or slightly lower) to those reported in previous studies ( $1.15$  and  $1.9 \text{ cm}^2/(\text{Vs})$  by Reischl, Mäkelä, Karch, and Nacid (1996),  $1.15$  and  $1.65 \text{ cm}^2/(\text{Vs})$  by Alonso, Kousaka, Nomura, Hashimoto, and Hashimoto (1997), and  $1.1$  and  $1.9 \text{ cm}^2/(\text{Vs})$  by Shimada et al. (2002) for positive and negative ions, respectively). The similarity in ion mobility values, regardless of the ion sources, suggests that the physical properties (i.e. size and mass) of the positive and negative ions generated by the carbon fiber charger are almost the same as those of the ions generated by the Kr-85 neutralizer.

### 3.2. Ion current and ozone concentration

Ion currents generated by the positive and negative ions from the carbon fiber ionizer at different voltages are shown in Fig. 4a. The airflow rate was 4 and 6 l/min for the positive and negative ionizer, respectively. A total airflow rate of 10 l/min

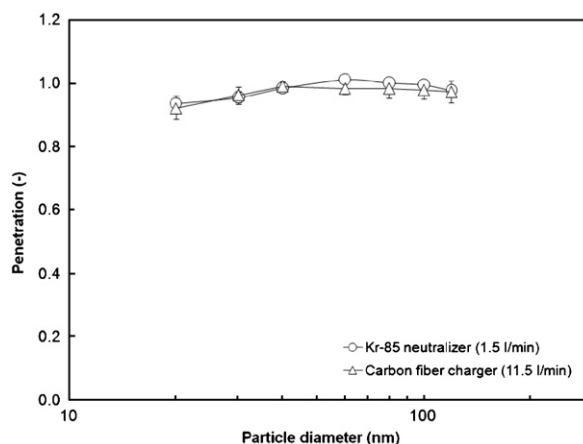


Fig. 5. Particle penetration through the carbon fiber bipolar charger.

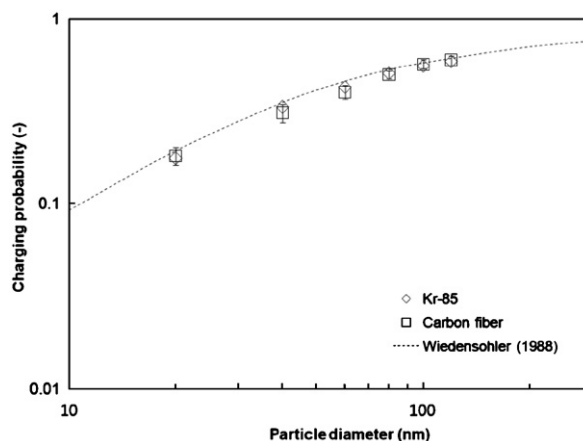


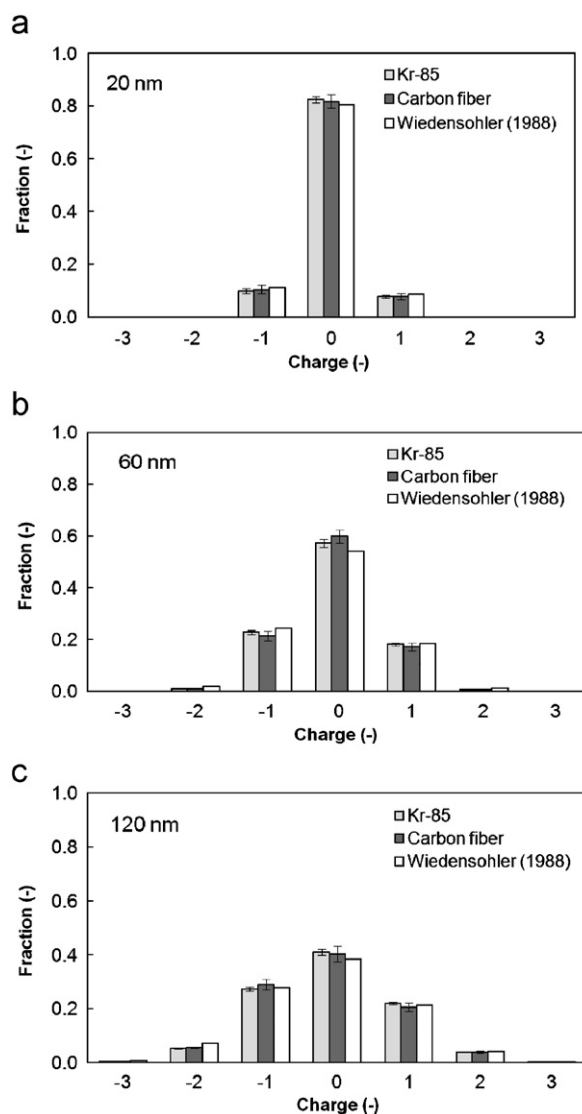
Fig. 6. Charge probabilities of particles neutralized by the carbon fiber bipolar charger and Kr-85 neutralizer for different particle sizes.

passed through the charging chamber and 3 l/min was introduced into the TSI aerosol electrometer. Positive ion currents were measured by varying the voltage applied to the positive ionizer while turning off the negative ionizer. The reverse process was followed to measure negative ion currents. The generation of positive and negative ions started at applied voltages of about +2.6 and –1.7 kV, respectively, and the ion concentrations increased sharply with the increasing voltage. The positive and negative ion values reached a near-saturation level at about +3.4 and –2.4 kV, respectively, due to a balance between the rates of generation and loss of ions in the SS cylinder at these increased voltages. The ion current for negative ions was lower than that for positive ions due to the greater loss of negative ions in the SS cylinder and transport tubes, which can be attributed to the higher mobility of negative ions. The total number concentrations for positive and negative ions measured at the outlet of the SS cylinders were higher than  $1.56 \times 10^6 / \text{cm}^3$ , which is the upper detection limit of the TSI 3068B electrometer (i.e. > 12.5 pA at 3 l/min).

The ozone concentrations were measured by means of a UV photometric ozone monitor (Model 1003-AH, Dasibi Environmental Corp., Glendale, CA) at the outlet of the charger and are shown in Fig. 4(a). The ozone concentrations at applied voltages less than –2.8 and +4.0 kV for negative and positive ionizers, respectively, were below the detection limit (2 ppb) of the  $\text{O}_3$  monitor. Detectable ozone concentrations were measured at –3.0 and at +4.5 kV and the concentration levels were about 10 ppb at –3.25 kV and 14 ppb at +5.0 kV, levels still negligible by the standards of most commonly used electrostatic chargers. It has been known that smaller diameter of a corona electrode and less oxidized material lead to lower generation of ozone (Nashimoto, 1988; Boelter & Davidson, 1997). Owing to the very small diameter of the carbon fibers, it is conceivable that the electric field at the tip of carbon fiber becomes high enough to generate corona discharge at a relatively low applied voltage compared to existing corona chargers, but still low enough to not generate high amounts of ozone.

To evaluate particle neutralization achieved by the new bipolar charger, the airflow rates through the ionizers were adjusted. Ion currents from the mixed stream of bipolar ions were measured at a voltage of +3.5 kV and an air flow rate of 4 l/min for the positive ionizer, and at a voltage of –2.5 kV and different air flow rates for the negative ionizer, and the results were shown in



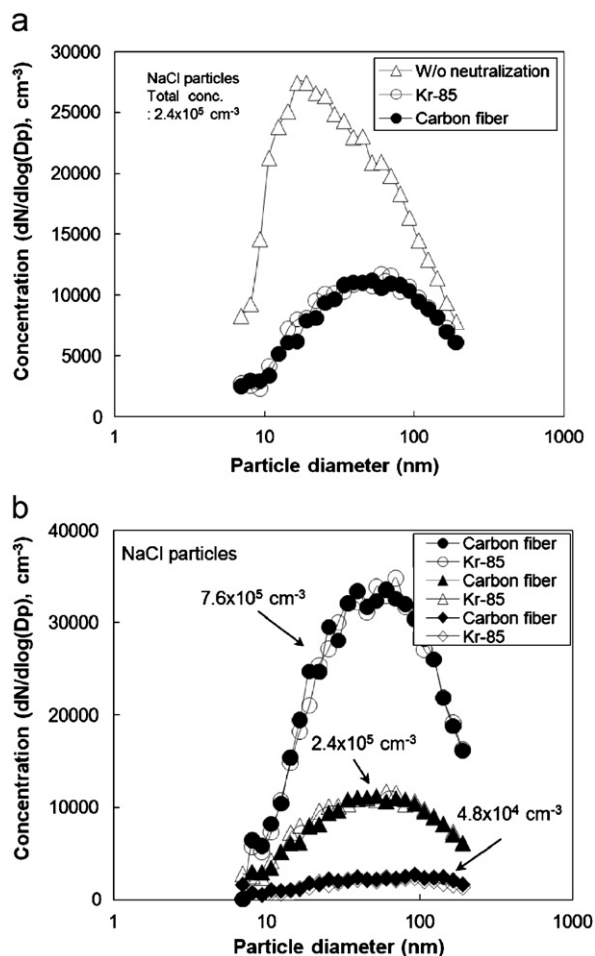


**Fig. 7.** Charge distributions of particles neutralized by the carbon fiber bipolar charger and Kr-85 neutralizer for different particle sizes: (a) 20, (b) 60 and (c) 120 nm.

**Fig. 4b.** Ion current of bipolar ions decreased with increasing airflow in the negative ionizer and changed to negative values at a flow rate of 7 l/min. The ion current of bipolar ions was about +100–+200 fA at a flow rate of 6 l/min in the negative ionizer, which was similar to that of the Kr-85 neutralizer (about +100 fA), even though it was not as stable as that obtained from the Kr-85 neutralizer. Based on the tests above, a voltage of +3.5 kV and an airflow rate of 4 l/min was used for the positive ionizer, and 2.5 kV and 6 l/min for the negative ionizer, as the optimum condition for neutralization of the carbon fiber bipolar charger. In this condition, ozone concentration was below the detection limit of the ozone monitor, as shown in Fig. 4a.

### 3.3. Particle penetration

Fig. 5 shows particle penetration through the carbon fiber bipolar charger for particles of different sizes. For measuring particle penetration, monodisperse particles were introduced into the carbon fiber charger, followed by a CPC (TSI 3022A). The particle penetration was determined as the ratio of the number concentrations detected by the CPC, with the carbon fiber charger turned on and off, respectively. The charger was operated in the optimum neutralization condition discussed earlier. Particle penetration greater than 92% was observed in the particle size range of 20–120 nm, nearly identical to that observed with Kr-85 neutralizer. This result indicates that electrostatic losses for particles larger than 20 nm are negligible in the carbon fiber charger.



**Fig. 8.** Size distributions of NaCl particles neutralized by the carbon fiber bipolar charger and Kr-85 neutralizer: (a) at particle concentration of  $2.4 \times 10^5 \text{ cm}^{-3}$  (including the one obtained without neutralization) and (b) at different particle concentrations.

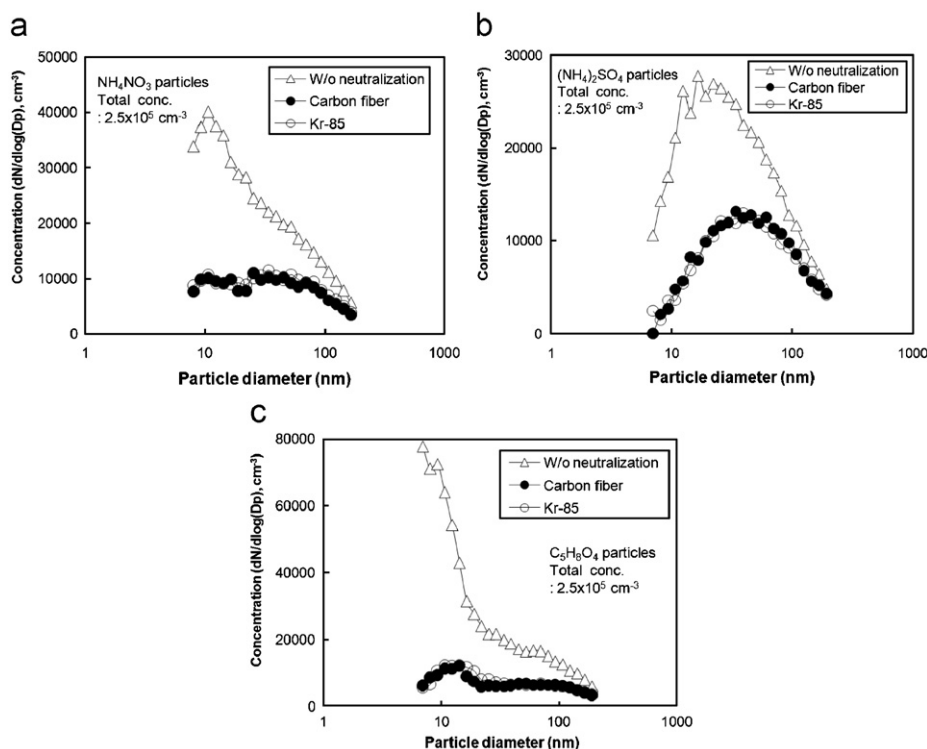
### 3.4. Charging probability

Fig. 6 shows the charging probability as a function of particle size for particles in the range 20–120 nm. The charging probability increases with the particle size because of the higher attachment of diffusive ions to the larger particles. The charging probability obtained by the carbon fiber charger was nearly the same as that by the Kr-85 neutralizer. The broken line in this graph indicates the empirical approximation of the Fuchs (1963) diffusion charging theory (Wiedensohler, 1988). The charging probabilities observed for both the carbon fiber charger and Kr-85 neutralizer were in good agreement with the theoretical predictions.

### 3.5. Charge distribution

Fig. 7 shows the charge distributions of particles observed for the carbon fiber charger and Kr-85 neutralizer, as well as the theoretical estimations by Wiedensohler (1988) for particles of size: (a) 20, (b) 60 and (c) 120 nm. Most of the 20 and 60 nm particles were singly charged, as expected. Fraction of negatively charged particles was slightly higher than positively charged, with the ratio of negative to positively charged particles being 1.2–1.4, due to higher mobility of negative ions, as shown in Fig. 3. Small fractions of doubly and even triply charged particles were found for 120 nm-sized particles. Charge distributions observed for the carbon fiber charger were similar to those of the Kr-neutralizer, as well as the theoretical predictions for particles of different sizes. These results indicate that the carbon fiber bipolar charger can bring submicron aerosol particles to the well-known stationary charge states as effectively as the Kr-neutralizer.





**Fig. 9.** Size distributions of various particles neutralized by the carbon fiber bipolar charger and Kr-85 neutralizer and comparison with that obtained without neutralization at particle concentration of  $2.5 \times 10^5 \text{ cm}^{-3}$ : (a) ammonium nitrate, (b) ammonium sulfate and (c) glutaric acid particles.

### 3.6. Size distribution

Fig. 8(a) shows the size distributions of NaCl particles measured by a DMA and CPC (i.e. SMPS system; Model 3936, TSI Inc., St. Paul, MN) after being neutralized by the carbon fiber bipolar charger and Kr-85 neutralizer, as well as without any neutralization. A similar size distribution, with an average size of about 62 nm and a geometric standard deviation of 2.3, was observed with both the carbon fiber bipolar charger and Kr-85 neutralizer. However, the size distribution without any neutralization had a much smaller mean size and a higher number concentration, because of considerably high initial charges on the particles generated by the nebulizer. These initially charged particles could be neutralized by the carbon fiber bipolar charger as effectively as the Kr-85 neutralizer.

Fig. 8b shows the size distributions of NaCl particles at different particle number concentrations, from  $5 \times 10^4$  to  $8 \times 10^5 \text{ particles/cm}^3$ . Both the carbon fiber bipolar charger and Kr-85 neutralizer resulted in similar size distributions, regardless of particle number concentration, which corroborates the effectiveness of the carbon fiber charger in neutralizing aerosols even at relatively high concentrations (i.e. nearly  $10^6 \text{ particles/cm}^3$ ) for most practical applications in aerosol researches.

Size distributions of various particle types, such as ammonium nitrate, ammonium sulfate and glutaric acid were measured after neutralized by the carbon fiber charger and Kr-85 neutralizer, as well as without neutralization, as seen in Fig. 9a–c. The generated particles were initially charged and thus displayed size distributions characterized by smaller particle sizes and higher number concentrations (when measured without neutralization) compared to the neutralized ones obtained by both chargers, which was in accordance with the result for NaCl particles shown in Fig. 8a. Glutaric acid particles had very high initial charges, and thus, their original size distribution shifted to even smaller sizes compared to other particle types, as shown in Fig. 9c. However, after neutralization using the carbon fiber charger, glutaric acid particles showed nearly the same size distribution as that observed for Kr-85 neutralizer. Therefore, the carbon fiber bipolar charger can be applied as an effective particle neutralizer, irrespectively of the initial charged states for particles of variable chemical composition.

## 4. Summary and conclusions

We developed a new carbon fiber bipolar charger to neutralize submicron aerosol particles. The performances of the charger, including particle penetration, charging probability, charge distribution and size distribution have been evaluated experimentally for submicron particles in the range of 20–120 nm. The experimental data were compared to those obtained for a Kr-85 radioactive

neutralizer as well as theoretical predictions. We found that the carbon fiber bipolar charger generated nearly negligible ozone and could neutralize submicron aerosol particles as effectively as commercial radioactive neutralizers at a flow rate of 1.5 l/min. The experimental results were in good agreement with those predicted theoretically. We thus believe that the carbon fiber bipolar charger can be an attractive alternative to radioactive neutralizers. A possible limitation of this technique is that the ion concentrations generated by the carbon fiber charger are somewhat more sensitive to changes in the operating conditions, such as applied voltage and airflow rate, compared to radioactive neutralizers. Therefore, this bipolar charger requires a relatively frequent monitoring of its ion balance, which is very crucial in assuring its operation under optimum neutralization conditions.

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